

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

#29

In re PATENT APPLICATION of

ESCH et al.

Group Art Unit: 1103

Rule 62 Continuation of  
Appln. No.: 08/594,327

Examiner: Hendrickson

Filed: June 6, 1997

FOR: PRECIPITATED SILICAS

\* \* \* \* \*

DECLARATION UNDER 37 CFR 1.132Hon. Commissioner of Patents  
and Trademarks  
Washington, D.C. 20231

Sir:

I, Udo Görl, a citizen of Germany, hereby declare and state  
as follows:

1. I hold a doctoral degree in chemistry from  
(academic degree and field)

University of WUERZBURG and have worked in the field of  
(Academic Institution)

Rubber Technology for more than 10 years. I  
(Specialty) (years of experience)

am an author on more than 10 scientific publications and  
(number)

a great number of presentations and patents  
(insert other professional experience if desired)

I currently hold the position of Manager Product Development Silicas  
(current title and place of  
and Silanes at Degussa AG, Rubber Chemicals and Pigments Division  
employment)

Applied Technology.

  
Dr. Udo Görl

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2. I am an Inventor on Patent Application No. 08/594,327. I am familiar with the Office Action of December 6, 1996 in the subject application, and with the presently pending claims, a copy of which is attached. In the Office Action, claims 1 and 7 were rejected under 35 U.S.C. § 102(b) or § 103 as allegedly lacking novelty or being obvious over Lagarde et al. (U.S. Patent No. 4,704,425).

3. Under my direct supervision, tests were conducted to compare the properties of the presently claimed compositions with those in the Lagarde patent.

4. Silicic acids Ultrasil 3370 GR (oven-dried) and Ultrasil 3380 GR (flash-dried) were tested in the recipe of U.S. Patent 4,704,425, column 14, as shown in attached Table 1. Both of these products are examples of the invention disclosed and claimed in the above-referenced patent application.

5. During the determination of the vulcanizate data the same test methods were used insofar as details were provided (see Table V of the present application). Attached Table 2 shows a comparison of the analytic data of the silicic acids in U.S. Patent 4,704,425 (column 13, table IV) with the data measured on

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the compositions of the present invention as recited in claims 1 and 7 ("93 200 FH").

6. Attached Table 3 compares the vulcanizate data from Table V of U.S. Patent 4,704,425 with the values of compositions of the present invention (93 200 FH). It is clear that the products of the invention have a markedly higher reinforcement behavior (modulus 300%) than the silicic acid from U.S. Patent 4,704,425. The products according to the invention also show advantages in DIN wear (abrasion loss). The remaining data (resistance to rupture, elongation to rupture, resistance to tear) is very heavily influenced by the modulus. Substantial advantages are seen with the compositions of the invention in respect to these characteristics.


7. It is clear from these comparative tests that the compositions according to the present invention are different from those of Lagarde et al., as described in U.S. Patent 4,704,425, and exhibit marked advantages over the compositions of Lagarde. Thus, the compositions are novel and not obvious from the Lagarde patent.

8. I declare further that all statements made on information and belief are believed to be true, and further that

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these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and such willful false statements may jeopardize the validity of the instant patent specification or any patent issuing thereon.

By



Date

4. Sept. 1997

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TABLE 1: Formulation

Buna 1509	100
Silica	50
PEG 4000	3
ZnO	3
Stearic acid	1
Octylated diphenyl amine (OCD)	2
MBTS	0,75
DOTG	1,6
Sulfur	2,5

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TABLE 2: Analytical Data

	Silicas according US 4,704,425		Silicas according 93 200 FH	
	Flash Dried	Oven Dried	Flash Dried	Oven Dried
CTAB [m <sup>2</sup> /g]	250	250	165	165
BET [m <sup>2</sup> /g]	260	260	170	180
pH -	4,7	4,9	6,5	6,3
DBP [ml/100g]	350	250	205	195

TABLE 3: Vulcanizate Data

	Silicas according US 4,704,425		Silicas according 93 200 FH	
	Flash Dried	Oven Dried	Flash Dried	Oven Dried
Modulus 100 % [MPa]	1,49	1,49	1,7	1,7
Modulus 300 % [Mpa]	2,25	2,53	3,7	4,0
Abrasion loss [mm <sup>3</sup> ]	159	154	142	140
Resistance to Rupture [MPa]	18,5	15,9	11,9	10,3
Elongation to Rupture [%]	803	821	550	510
Resistance to Tear [N/mm]	>40	>40	30	30

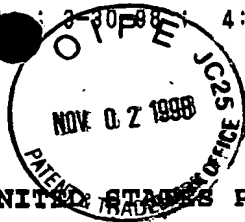
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Currently pending claims:

1. Precipitated silica having the following physico-chemical properties:

BET surface area	35 to 350 m <sup>2</sup> /g
BET/CTAB surface area ratio	0.8 to 1.1
Pore volume, PV	1.6 to 3.4 ml/g
Silanol group density, ml of NaOH consumed in raising pH to 9	6 to 20 ml
Average aggregate size	250 to 1500 nm
CTAB surface area	30 to 350 m <sup>2</sup> /g
DBP value	150 to 300 ml/100 g
V <sub>2</sub> /V <sub>1</sub> by Hg porosimetry	0.19 to 0.46
DBP/CTAB	1.2 to 3.5

7. The precipitated silica according to claim 1, wherein the DBP/CTAB ratio is 1.2 to 2.4.



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re PATENT APPLICATION of

ESCH et al.

Group Art Unit: 1754

Appln. No.: 08/870,591

Examiner: Hendrickson

Filed: June 6, 1997

FOR: PRECIPITATED SILICAS

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DECLARATION UNDER 37 CFR 1.132

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and Trademarks  
Washington, D.C. 20231

Sir:

I, Udo Görl, a citizen of Germany, hereby declare and state  
as follows:

1. I hold a doctoral degree in chemistry from the  
UNIVERSITY OF WUERZBURG and have worked in the field of  
Rubber Technology for more than 10 years. I am an author on more  
than 10 scientific publications and a great number of  
presentations and patents. I currently hold the position of

Manager Research & Development Powder Rubber Technology - PKV GmbH, D-45764 Marl  
(current title and place of employment) Germany

2. I am an Inventor on Patent Application No. 08/594,327.  
I am familiar with the Office Actions of December 6, 1996,  
September 29, 1997 and June 8, 1998 in the subject application,  
and with the presently pending claims, a copy of which is  
attached. In the Office Actions, claims 1 and 7 were rejected

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under 35 U.S.C. § 102(b) or § 103 as allegedly lacking novelty or being obvious over Johnson et al. (U.S. Patent No. 4,681,750).

3. Under my direct supervision, tests were conducted to compare the properties of the presently claimed compositions with those in the Johnson patent ('750).

4. Silicic acid Ultrasil 3380, according to the present invention, and LV 6589, which was manufactured in accordance with the data contained in the '750 patent, were compared using a high silicic acid-filled PKU tread mixture based on L-SBR/BR.

5. The attached tables show a comparison of the analytic data and rubber technology characteristics of Ultrasil 3380 and LV 6589. Substantial advantages are seen with the composition of the invention in respect to these characteristics. This is especially clear in the strength values and in the modulus, but also in the ball rebound (corresponding to lower rolling resistance) and in the  $\tan \delta$  at  $0^\circ$  (higher values signifying better wet skid resistance properties). The evaluation of the dispersion using a 10-point scale (Phillips method) shows that 3380 is two steps better than LV 6589. Most of these application technology advantages are associated with the lower Sears number

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of the present invention, which stems from the different manner of precipitation.

6. It is clear from these comparative tests that the compositions according to the present invention are different from those of Johnson et al., as described in U.S. Patent 4,681,750, and exhibit marked advantages over the compositions of Johnson et al. Thus, the compositions are novel and not obvious from the Johnson patent.

7. I declare further that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and such willful false statements may jeopardize the validity of the instant patent specification or any patent issuing thereon.

By



Date

15<sup>th</sup> October, 1998

Comparison of 3380 with LV 6589

## 1) Analytical Data

	CTAB m <sup>2</sup> /g	pH	DBP g/100g	Sears value V2[ml]
3380	165	6.5	278	17.5
LV 6589	163	6.3	312	20.6

## 2) Rubber Technology

Test Method	Unit	3380	LV 6589	Norm
ML 1+4	-	73	77	DIN 53523/3
Strength	Mpa	13.2	11.9	DIN 53504
300% modulus	Mpa	11.7	11.0	DIN 53504
Break Stretch	%	330	320	DIN 53504
Break Energy	$\tau$	59.9	52.0	DIN 53504
Hardness	-	66	67	DIN 53505
Ball Rebound 60°C	%	61.3	58.1	ASTM D 5308
DIN Abrasion	mm <sup>3</sup>	58	61	DIN 53516
Loss Angle tan $\delta$ (0°C)	-	0.473	0.434	DIN 53513
Dispersion (Philips)	-	9	7	ISO/DIS 11345

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Pore volume, PV	1.6 to 3.4 ml/g
Silanol group density, ml of NaOH consumed in raising pH to 9	6 to 20 ml
Average aggregate size	250 to 1500 nm
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DBP value	150 to 300 ml/100 g
V <sub>2</sub> /V <sub>1</sub> by Hg porosimetry	0.19 to 0.46
DBP/CTAB	1.2 to 3.5

7. The precipitated silica according to claim 1, wherein the DBP/CTAB ratio is 1.2 to 2.4.